Relationship between Refractive Index and Density and Consistency of Mixing Rules

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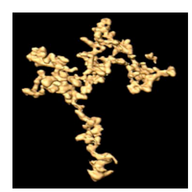
(Please see “Liu and Daum, J. Aerosol Science, 39, 974-986, 2008” for detail)

- *Is there a index-density relationship and why?*
- *Mixing rules and consistency with such a relationship*



Santa Fe
27 Feb 2009

Motivation

*** <u>General:</u> Both refractive index (optical properties) and mass density (aerosol dynamics) are needed to improve aerosol measurements and modeling such as GCMs.**

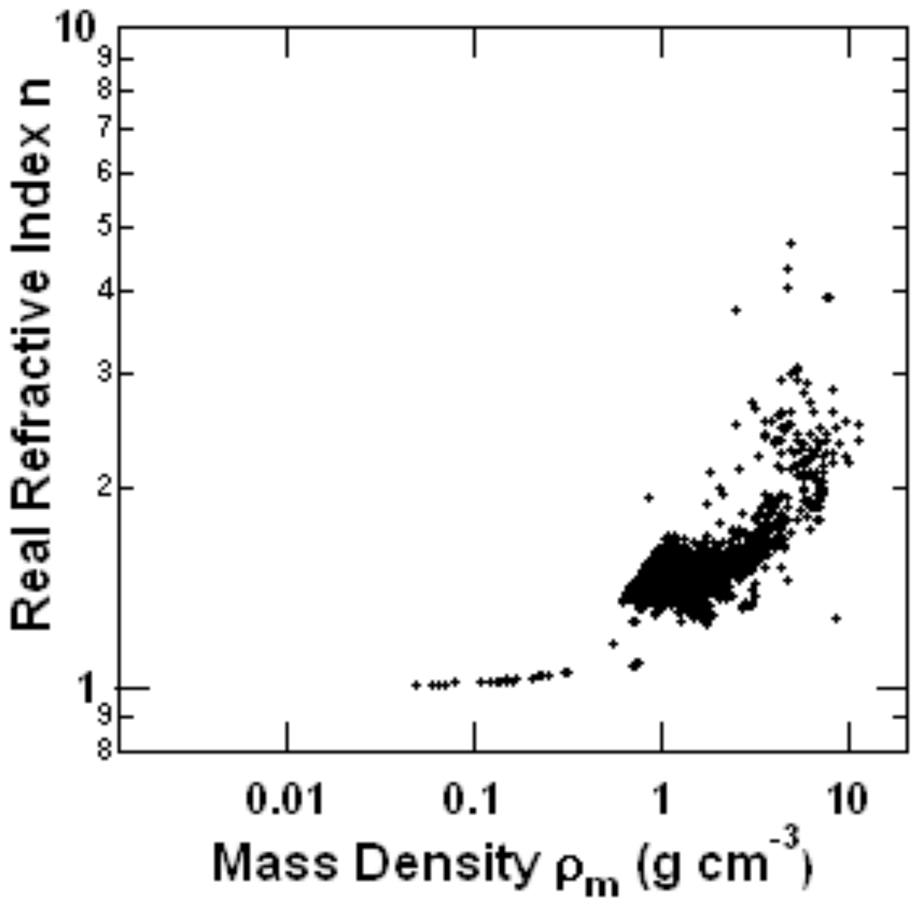
*** <u>ASP Specific:</u>**

- Last meeting: Improving mixing rules was recommended as a priority, which was a direct motivation for submission of this paper.

- This meeting (S. Martin this am): Mixing rules continue to be a high priority, and "optical dynamics" is emphasized.

Calling for a unified view/treatment of refractive index and density!

Empirical Evidence for the Refractive Index-Density Relationship



Each dot denotes a material; a total of over 4000 materials are shown, including organics, inorganics, gases, liquids, solids. Data are from various sources, especially www.knovel.com (thanks to Dr. S. E. Schwartz for pointing us to this electronic data library .

This large collection of data clearly demonstrates that refractive index and mass density are positively correlated to each other in general.

Physical Theory for the Refractive Index-Density Relationship

• The theoretical basis was largely established in the late 19th and early 20th century in other fields ---- a brief revisit here.

• Qualitative physics: A denser material tends to have a larger refractive index because more electric dipoles will be activated when exposed to an electric field.

• Quantitative theory: Lorentz-Lorenz relation

$$r \equiv \frac{n^2 - 1}{n^2 + 2} = \frac{N_A \alpha}{3M} \rho_m$$

n = Refractive index; ρ_m = Mass density;
N_A = Avagadro's number; M – Molecular weight
α– Molecular polarizability
r = Specific refractive index

The Lorentz-Lorenz relation has important implications for developing self-consistent mixing rules used to estimate effective density and effective refractive index of ambient aerosols.

Consistent Mixing Rules for Ambient Aerosol Particles

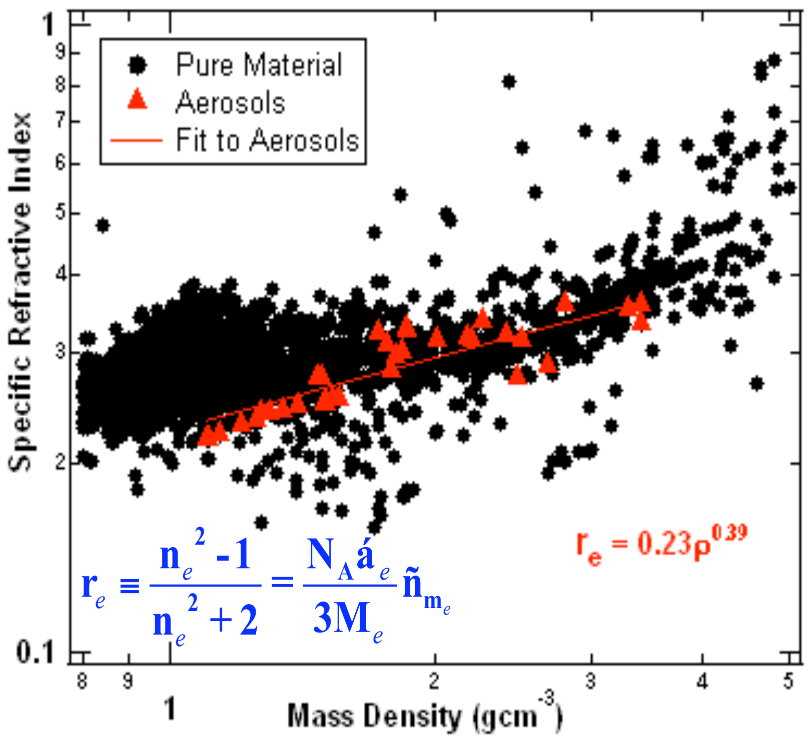
Consistency demands that the effective properties of a mixture satisfy the Lorentz-Lorenz relation as well. That is:

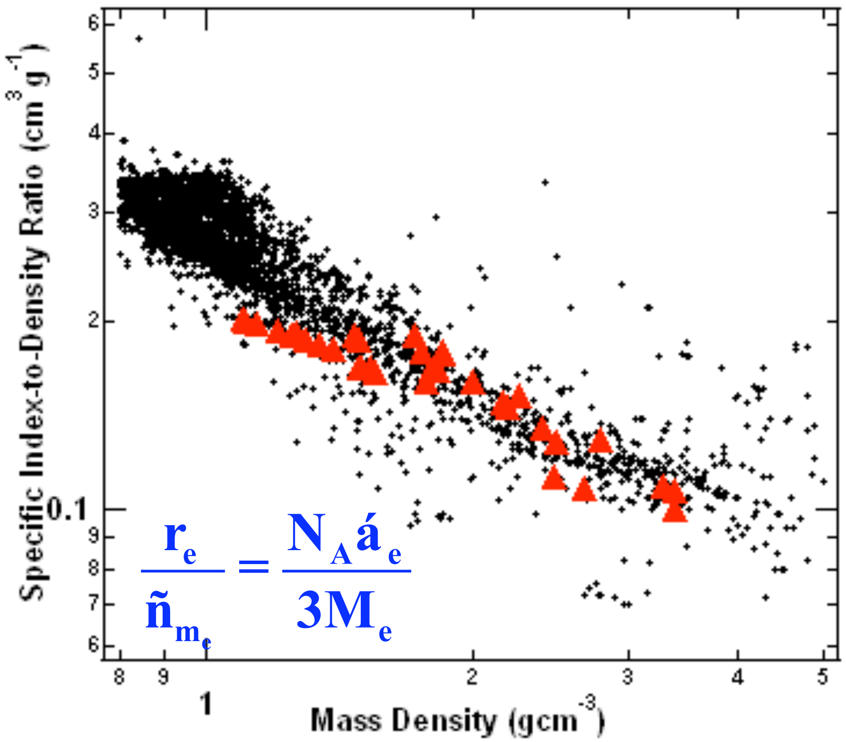
$$r_e \equiv \frac{n_e^2 - 1}{n_e^2 + 2} = \frac{N_A \acute{a}_e}{3M_e}\tilde{n}_{m_e} \qquad \Longleftrightarrow \qquad R_e \equiv \frac{\left(n_e^2 - 1\right)M_e}{\left(n_e^2 + 2\right)\tilde{n}_{m_e}} = \frac{N_A \acute{a}_e}{3}$$

(r = specific refraction; R = molar refraction)

- ***The Lorentz-Lorenz mixing rule*** **for calculating effective refractive index is consistent with the Lorentz-Lorenz relation.**
- ***The Molar fraction mixing rule*** **is equivalent to the Lorentz-Lorenz mixing rule.**
- ***The volume mixing rule*** **for calculating effective refractive index is approximately consistent with the Lorentz-Lorenz relation for quasi-homogeneous mixture where refractive indices of different components do not differ much.**

Application to Ambient Aerosols and Implications for Future Research





The exponent of 0.39 $<$ 1 indicates that α_e/M_e or r_e/M_e decreases with increasing density, as shown in the right plot.

These results suggest the need to simultaneously measure refractive index, mass density and polarizability, …., recall Peter's talk on